

**STUDIES OF ATOMIC SPECTRA
AND TRANSITION PROBABILITIES**

**BY
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STUDIES OF ATOMIC SPECTRA AND TRANSITION PROBABILITIES

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AKADEMISK AVHANDLING

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1. LIST OF PUBLICATIONS.

The 16 published papers in this thesis can be naturally divided into three groups: spectral studies, studies of atomic lifetimes, and investigations of the beam-foil excitation mechanism. In order to structure this thesis the second group has been further divided into lifetime analyses not requiring extensive cascade corrections and those that do.

For the papers with several authors, we have generally used the principle that the first name refers to the person primarily responsible for the planning and analysis of the experiment.

GROUP 1. SPECTROSCOPIC STUDIES.

1.1 Beam - Foil Studies of Multiply Ionized Oxygen.

B.Denne, L.Engström, S.Huldt, J.O.Ekberg, L.J.Curtis,
K.Ishii, E.Veje and I.Martinson.
Physica Scripta 21, 151 (1980).

1.2 The Spectrum of Six Times Ionized Fluorine.

L.Engström
Physica Scripta 29, 113 (1984).

1.3 Transitions in Multiply Excited F VI and F VII.

I.Martinson, B.Denne, J.O.Ekberg, L.Engström, S. Huldt,
C.Jupén, U.Litzén, S.Mannervik and A.Trigueiros
Physica Scripta 27, 201 (1983)

1.4 Extended Analysis of Cr IV.

J.O.Ekberg and L.Engström.
Physica Scripta 25, 611 (1982).

GROUP 2. STUDIES OF ATOMIC LIFETIMES.

2.1 ANALYSES NOT REQUIRING EXTENSIVE CASCADE CORRECTIONS.

2.1.1 Lifetime Measurements for some $\Delta n = 0$ Transitions in Cl XIII - Cl XV.

K.Ishii, E.Alvarez, R.Hallin, J.Lindskog, A. Marelus,
A. Pihl, R.Sjödin, B.Denne, L.Engström, S.Huldt and
I.Martinson.

Physica Scripta 18, 57 (1978).

2.1.2 Experimental Transition Probabilities for Intercombination Lines in N IV, O V and F VI.

L.Engström, B.Denne, S.Huldt, J.O.Ekberg, L.J.Curtis,
E.Veje and I.Martinson.

Physica Scripta 20, 88 (1979).

2.1.3 Lifetimes of the $1s2p\ ^3P_0$ and 3P_2 Levels in F VIII

L.Engström, C.Jupén, B.Denne, S.Huldt, Weng Tai Meng,
P.Kaijser, U.Litzén and I.Martinson.

J.Phys.B 13, L143 (1980).

2.1.4 Transition Probabilities for Allowed and Forbidden Transitions From the $1s2p\ ^3P$ Levels in He-like O VII and F VIII.

L.Engström, C.Jupén, B.Denne, S.Huldt, Weng Tai Meng,
P.Kaijser, J.O.Ekberg, U.Litzén and I.Martinson.

Physica Scripta 22, 570 (1981).

2.2. LIFETIME ANALYSES INVOLVING CASCADE CORRECTIONS THROUGH THE
ANDC METHOD

- 2.2.1 CANDY, a Computer Program to Perform an ANDC Analysis
of Cascade Correlated Decay Curves.

L.Engström

Nucl.Instr.Meth. 202, 369 (1982)

- 2.2.2 Lifetimes of the $2s2p\ ^1P$ Level in Be-like N IV and O V
Obtained from Beam-Foil Experiments and Extensive Cascade
Analyses.

L.Engström, B.Denne, J.O.Ekberg, K.W.Jones, C.Jupén,
U.Litzén, Weng Tai Meng, A.Trigueiros and I.Martinson.
Physica Scripta 24, 551 (1981).

- 2.2.3 Accurate Experimental Lifetimes of Excited Levels in
Sodiumlike Sulfur, S VI.

J.O.Ekberg, L.Engström, S.Bashkin, B.Denne, S.Huldt,
S.Johansson, C.Jupén, U.Litzén, A.Triguerios and
I.Martinson, Physica Scripta 27, 425 (1983)

GROUP 3. STUDIES OF THE BEAM-FOIL EXCITATION MECHANISM.

- 3.1 Observation of Strong Orientation Effects for Levels in Highly Ionized Oxygen.

S.Huldt, L.J.Curtis, B.Denne, L.Engström, K.Ishii and I.Martinson.

Phys.Lett. 66A, 103 (1978).

- 3.2 Relative Level Populations in Beam-Foil-Excited C IV, N V, O VI and F VII.

B.Andresen, B.Denne, J.O.Ekberg, L.Engström, S.Huldt, I.Martinson and E.Veje.

Phys.Rev. A23, 479 (1981).

- 3.3 Study of the Beam-Foil Excitation Mechanism with the Use of Chlorine Projectiles, 2 - 10 MeV.

C.Jupén, B.Denne, J.O.Ekberg, L.Engström, U.Litzén I.Martinson, W.Tai-Meng, A.Trigueiros and E.Veje.

Phys.Rev. A26, 2468 (1982)

- 3.4 Relative Level Population in S VI after Beam-Foil Excitation, Obtained from ANDC Analyses of Measured Decay Curves.

L.Engström

Physica Scripta 28, 68 (1983)

- 3.5 Dissociation of CN^+ into Atomic Excited States after Acceleration in a Tandem Accelerator.

H.G.Berry, L.Engström, R.Hellborg, S.Huldt, C.Jupén, A.Trigueiros and I.Martinson.

Phys.Letters 97A, 187 (1983)

2. INTRODUCTION.

Knowledge of the energy level structure and spectra of atoms and ions together with the decay properties (lifetime and transition probabilities) of the excited states is of tremendous importance, not only within atomic physics but also in many other fields of science and, in the long run, also from a technological point of view.

Atomic physics is a basic science which evolves through the interaction between theory and experiment, and a large number of different theoretical methods have been developed for the calculation of atomic structure and transition probabilities. The former problem is thoroughly discussed by e.g. Cowan [1] and Hibbert [2], whereas Crossley [3] has given a recent review of the calculation of atomic transition probabilities.

An example of the interplay between theory and experiment is provided by the study of atomic lifetimes. When lifetime data for levels in multiply ionized atoms became available through beam-foil experiments in the 1960's it was often found that the calculated lifetimes were in clear disagreement (a factor of 2-3) with the experimental results, even for comparatively simple atomic systems [4]. This stimulated work leading to a substantial improvement of the theoretical results and, as we shall see in section 6, the situation is now in some cases reversed. Thus, refined calculations for low-lying levels in few-electron systems have shown that the earlier beam-foil experiments gave systematically too long lifetimes (10-20%) due to incomplete corrections for cascades.

In the calculation of energy levels and transition probabilities the major problem for low-Z ions is to account for the

correlation between the electrons, whereas for heavier atoms relativistic effects become important [1,2]. For one and two electron systems high-precision measurements can also be used to test quantum electrodynamic (QED) contributions to the transition energies [5,6].

Outside the field of atomic physics the major users of atomic data are probably astrophysicists and people working in plasma physics and fusion research. In both cases wavelength data are needed for the identification of the various chemical elements present in astronomical object and laboratory plasmas. Once the radiating system (atom and charge state) has been identified, transition probabilities are needed in order to determine its abundance. This knowledge can then be used to study the composition and dynamics of the source. Many astronomers use for example a comparison between an observed spectrum and one obtained from a model calculation to study processes in stellar atmospheres [7]. A prerequisite for such investigations is the existence of a large atomic data base. The intensive efforts to achieve controlled fusion has also greatly increased the demands for atomic data, particularly for highly ionized atoms, since spectroscopic techniques are used in the diagnostics of the plasma [8].

This important collaboration between atomic physics, astrophysics and plasma physics was the topic of a symposium in Lund in August 1983, and for further details and examples the reader is referred to the proceedings of that meeting [9]. Also, for a thorough discussion of experimental methods and results concerning the study of highly ionized atoms of astrophysical and plasma-physical relevance the review by Martinson [10] should be consulted.

3. THE BEAM-FOIL METHOD.

Since the experiments reported in all but one (paper 1.4) of the papers in this thesis are performed with the beam-foil technique this section will be devoted to a description and discussion of this experimental method.

The pioneering work on the use of a foil-excited ion beam for atomic physics experiments was done in the early 1960's by Kay [11] and Bashkin [12]. Since then the method has been extensively used in many laboratories, primarily as a unique way of measuring atomic lifetimes in multiply ionized atoms. In addition, the foil-excited beam may also be used as a light source for spectroscopic studies, where its special properties make it a valuable complement to other techniques for the generation of atomic spectra.

There exist several review articles on beam-foil spectroscopy. A few of the more recent ones are given in Refs. 13-16. In addition, a thorough discussion of the method, including technical details on the experimental setup, ways to obtain narrow spectral lines and the numerical analysis of lifetime measurements can be found in [17].

The principal setup of a BFS experiment is shown in Fig 1. An accelerator provides a beam of fast ions which, after charge-state momentum analysis, passes through a thin foil (usually made of carbon with a thickness of a few hundred ångström) placed on a movable carriage within the target chamber. The beam is finally directed to a Faraday-cup where the beam-charge is collected and used to normalize the measurements against beam fluctuations.

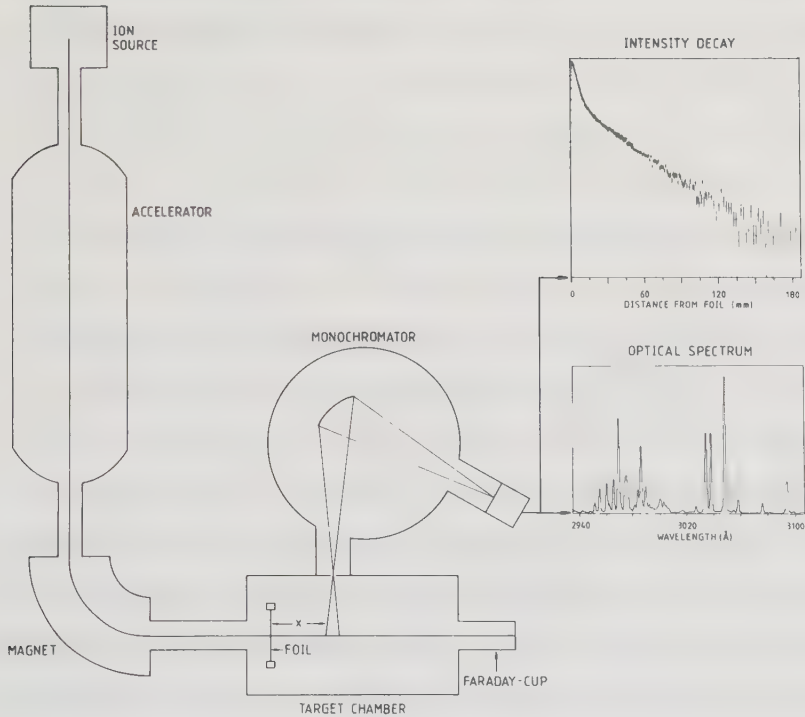


Fig 1. The principal setup of a beam-foil experiment.

The interaction between the ions and the foil atoms results (normally) in a significantly increased degree of ionization of the beam. Actually, the ions will be distributed on a number of charge states. The mean and width of these distributions depends only on the velocity of the incoming ions (for a given projectile element and a given foil material of sufficient thickness) [18].

As an example, a 5 MeV beam of fluorine gives the charge states F^{+3} - F^{+7} with the relative intensities 10, 50, 100, 50 and 10 respectively, regardless of the incoming charge state.

Moreover, a sizable fraction of the emerging ions will not be in their energetic ground state, but instead distributed without any apparent restrictions on the infinite number of possible

excited states. As an example, in a recent experiment with S ions Kanter et al. [19] found evidence for the excitation of levels with n -values as high as in the range 250 - 650. No comprehensive theory exists at the moment on exactly how these distributions look nor on the details of the beam-foil interaction itself. This interesting problem is discussed in more detail in section 7 of this thesis.

Fig 1. illustrates also the two major types of experiments performed: spectroscopic studies and measurements of atomic lifetimes.

In the former case the foil is held fixed as close to the entrance slit of the monochromator as possible and the light emitted by the ions is registered by a photomultiplier after wavelength dispersion. Due to the low light intensities the photomultiplier is operated in a photon-counting mode. This ensures also a very linear intensity scale over a large dynamical range.

As a light source for spectroscopic studies the beam-foil interaction offers several important and rather unique advantages.

1. Almost any ionization stage of almost any atom can be studied.
2. Lines belonging to different ionization stages are easily identified.
3. Unselective excitation. Many different levels are excited and, in comparison with other light sources, larger populations of hydrogenic and multiply excited states are obtained.
4. Pure spectra, i.e. no impurity lines are present.
5. Undisturbed decay, i.e. the lines are not affected by the Stark effect, collisional broadening or radiation trapping.

The method has also some disadvantages, which limit the ultimate spectral resolution and wavelength accuracy obtainable.

6. Low light intensities.

7. High velocity of the light emitting ions.

To achieve the first point one has to consider all available accelerators and ion-sources in the world. Then it is possible to cover energies up to several hundred MeV for any element, and according to the discussion above (velocity dependent charge state distributions) most charge states can be produced. Also the 4:th point is related to the use of an accelerator since magnetic analysis of the ions ensures an isotopically pure beam. However, with single-stage accelerators without any prestripping of the ions before the analysing magnet, molecules may present a problem of momentum coincidences.

Thus, the recorded spectra contain only lines from a given element. But since the beam-foil interaction produces several charge states after the foil it is vitally important to be able to assign the observed lines to a specific ion. This is easily accomplished by comparing spectra recorded at different ion energies. This is illustrated in Fig 2, where part of a fluorine spectrum recorded at 3 and 4.5 MeV is shown. Note the clear difference in the relative intensities of lines belonging to different charge states! To ensure accurate assignments of the lines belonging to a particular charge state, especially for heavier ions, it is usually necessary to use 3 or 4 different energies.

Points 5 and 6 are directly related to the low particle density in the beam. At the present level of accuracy, this is the most serious limitation of the method since it necessitates the use of

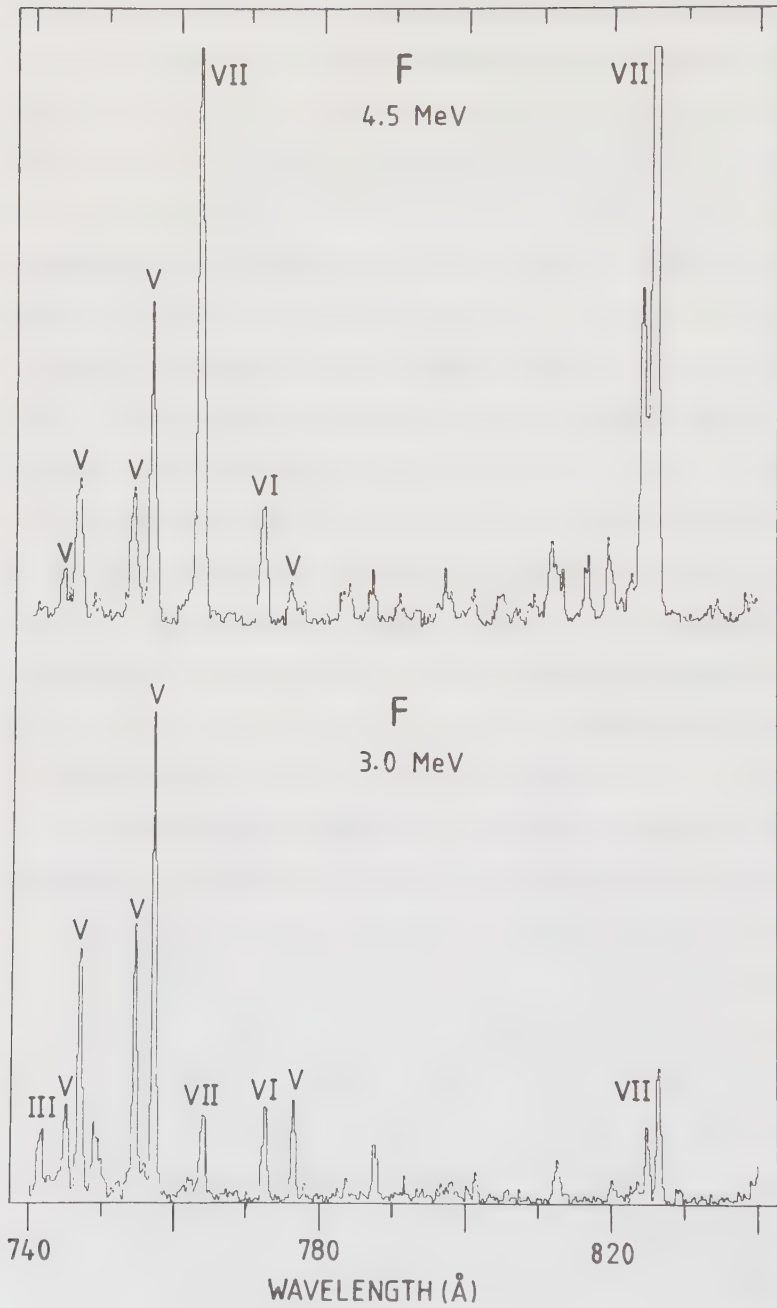


Fig 2. Part of a fluorine spectrum recorded at 3 and 4.5 MeV beam-energy.

faster spectrometers with wider slits than normally used for spectroscopic studies. Still, linewidths below 1 Å can routinely be obtained in studies covering large spectral regions (several thousand ångströms), and, substantially better resolution is possible within reasonable data-collection times and with an acceptable signal-to-noise ratio over more limited wavelength intervals. Furthermore, with the use of multichannel detectors (microchannel plate or diode array) the data-collection efficiency can be drastically increased [20,21].

The last characteristic of the beam-foil source (fast moving light source) requires careful consideration. First of all, observations must be made at an angle of 90 deg. to the beam in order to avoid Doppler shift of the lines. But even then, the fact that light is collected not from a point but from a section of the beam (see Fig 1.) leads to a large Doppler broadening of the lines. However, Stoner and Leavitt [22] have shown that this effect can be cancelled in the first order approximation by increasing the length of the light path within the spectrometer for example by moving the exit slit away from the grating. This technique is referred to as refocusing. Other methods of refocusing have also been developed by Bergkvist [23] and by Jelley et al. [24]. Because of these Doppler effects an accurate absolute wavelength scale can only be established with in-beam reference lines. That is, the spectrum must contain a sufficient number of accurately known lines with a reasonably uniform wavelength distribution.

Despite the rather lengthy and detailed discussion above on the use of the beam-foil technique as a source of atomic spectra it should be stressed that the most unique quality of the method is its time resolution. Thus, for multiply ionized atoms it is the

only general experimental technique available for the determination of atomic lifetimes.

The lifetime of an excited level is measured by recording the intensity of a spectral line as a function of the distance between the foil and the entrance slit of the monochromator. (This distance is marked x in Fig 1.) In an ideal case, the observed light intensity $I(x)$ shows a simple exponential decay:

$$I(x) = I(0) e^{-x/v\tau}$$

where v is the beam velocity and τ is the lifetime (defined as the time required for the intensity to be reduced by a factor $1/e$) of the upper level. The lifetime is then determined by numerically fitting the above expression to the observed decay curve. The lifetime τ of a level i is also given by the relation

$$\tau^{-1} = \sum_k A_{ik}$$

where A_{ik} is the transition probability for a spontaneous decay from level i to level k . See appendix 3.

Because of the unselective excitation of a large number of levels (point 3 above) the one under investigation will be constantly repopulated from higher-lying levels. This process is called cascading. Thus, in most real situations the simple one-exponential expression above must be replaced by a sum of exponential functions, which severely complicates the analysis. An example of such a multiexponential decay is shown in Fig 1. If one of the upper levels has a lifetime shorter than the primary one, then this sum will also contain a negative term, a so called growing-in cascade. The problem of analysing such multiexponential decays will be discussed in section 6.

4. THE EXPERIMENTAL FACILITY IN LUND.

The beam-foil facility at the University of Lund has been thoroughly documented in [25-27] (hardware) and [28,29] (software) and the most essential information is of course also given in the papers. Thus, this section gives only a brief review of the subject together with some comments on ongoing improvements. For a description of the setup at the Tandem Accelerator Laboratory in Uppsala (used in paper 2.1.1) the reader is referred to Hallin et al. [30].

The Lund Pelletron accelerator was manufactured by National Electrostatics Corporation and is of the tandem type with a maximum terminal voltage of 3 MV. In all experiments reported here a Duo-plasmatron ion source has been used. During 1983 it was, however, complemented with a sputtering source from Danfysik which will substantially increase the number of ions available for experiments [31]. A very detailed description of the accelerator is given by Hellborg and Håkansson [32].

The target chamber which has been in operation since 1978, holds 20 foils. The minimum displacement of a foil during a decay curve measurement is $1/16$ mm, whereas the decay can be followed over approximately 200 mm. During the last few years a new target chamber has been designed and built at our institute, Engström and Håkansson [33], and is scheduled for operation towards the end of this spring. The main improvement in the new design is the possibility of having three spectrometers, covering the 50 - 10000 Å wavelength region, simultaneously viewing the beam under optimum conditions. The chamber with two spectrometers is shown in Fig 3. The third instrument which will view the beam from below, is not shown. Further details are given in [33].

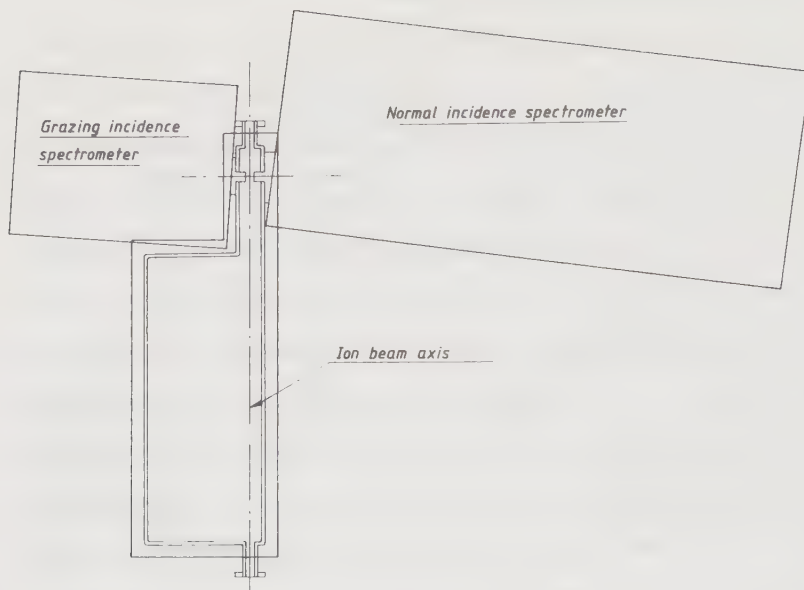


Fig 3. The new experimental setup. Only two of the three spectrometers are shown.

The spectrometer that has been used in most of the reported investigations is a Minuteman 310 NIV 1 m normal incidence vacuum monochromator. The original entrance slit assembly of this instrument has been redesigned and the new slit is now placed inside the target chamber. This arrangement increases the spatial (time) resolution for lifetime measurements which is important particularly when studying fast decays. A total length of 0.3 mm of the beam is normally seen, but this can be reduced by masking the grating as was done in paper 2.2.3. For studies involving wavelengths above 2000 Å two other spectrometers, optically coupled to the chamber, have also been used: a 1 m McPherson 2051 (papers 1.1, 2 1.2 and 3.2) and a 0.35 m McPherson EUE-700 (paper 3.1).

The last two monochromators are of the Czerny-Turner type and refocusing was achieved by moving the last concave mirror. According to [22] the displacement of either the grating (in a normal incidence spectrometer) or the last mirror (Czerny-Turner mount) is given by

$$\Delta = v \lambda / 2 c k$$

where v is the beam velocity, c the speed of light, λ the wavelength and k the inverse dispersion ($\text{\AA}/\text{mm}$). Thus, refocusing by this technique can only be accomplished for one particular wavelength, although the line widths increase very slowly for other wavelengths. However, for a scanning normal incidence monochromator it turns out that (almost) completely wavelength-independent refocusing can be done, if instead of moving the grating a fixed distance the focusing cam of the instrument is placed at an angle to its position for a stationary source. The angle will then introduce an additional displacement of the grating proportional to the wavelength, just as required according to the formula above. This technique was implemented on the Minuteman monochromator.

Also in the area of spectrometers there is intensive development going on at our institute. Thus, Hutton and Huldt [34] are designing a new spectrometer utilizing an echelle grating and optimized for use with the beam-foil source for wavelengths above 2000 $\text{\AA}$. This instrument is intended to become the third one on the new target chamber, thus completing a very powerful experimental facility.

The signal processing system is shown in Fig 4. The computer is a Computer Automation ALPHA-LSI 2/20 and most of the electronics are standard NIM modules. The stepmotor and scaler control units as well as the interface between these and the computer have been designed by Huldt and are documented in [25,26]. In the new setup

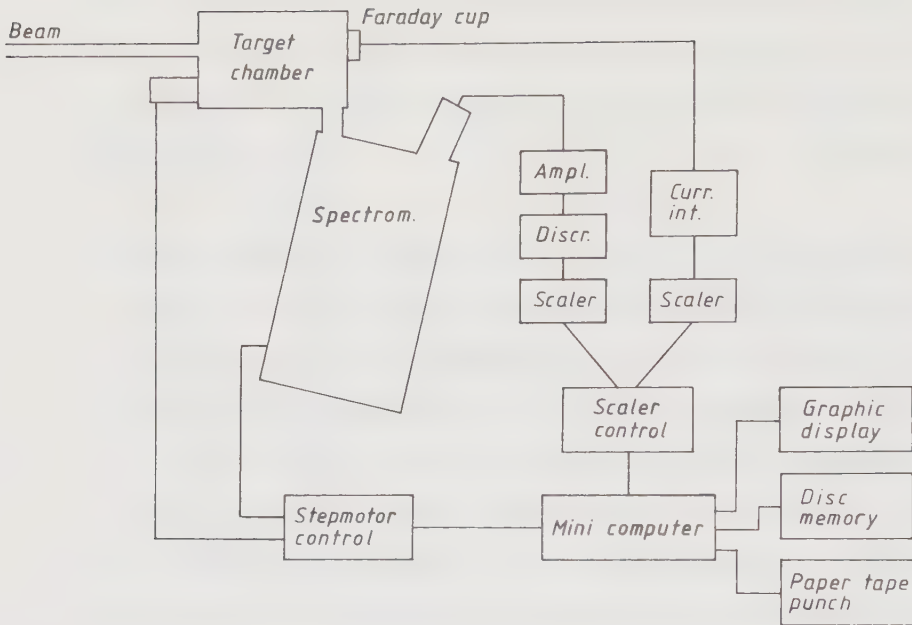


Fig 4. The signal processing system of the present experimental setup.

a CAMAC based system will be used in combination with a much more powerful computer [35].

The final part of our facility is of course the software needed to control the data-collection and to perform the analyses of the experimental material. These programs are written and thoroughly documented by Engström and Huldt [28] (data-collection) and by Engström [29] (analysis). A list of the most important programs together with a very brief description of their purposes is given in appendix 1.

5. SPECTRAL STUDIES.

As a suitable introduction to this section we quote R.D.Cowans description of a term analysis. "The problem is rather like that of trying to put together the pieces of a complicated jigsaw puzzle when the pieces never fit exactly, some pieces fit spuriously, some critical pieces are missing, and there are pieces present that belong to one or more entirely different puzzles". (Ref. 1 p. 27).

Papers 1.1 - 1.3 report studies of atomic energy levels and spectra where the BF method has been used to obtain the spectrograms, whereas in paper 1.4 a sliding-spark source has been utilized.

The first three papers illustrate how the special properties of the beam-foil interaction (particularly points 3 and 5 mentioned in section 3) can be used to complement other light sources in the studies of atomic structure. Thus, in papers 1.1 and 1.2 we have emphasized the study of transitions between levels with high n and ℓ quantum numbers in the Li-like spectra O VI and F VII, whereas paper 1.3 reports a study of multiply excited levels in Li- and Be-like F.

Papers 1.1 and 1.2.

In 1979 Edlén [36] published a major work on the Li I iso-electronic sequence in which accurate semi-empirical formulae are given that predict the complete $1s^2n\ell^2L$ energy level structure for ions with Z up to around 30. With the main intention of testing these predictions, we performed a study of the spectra O VI and F VII.

In the fluorine case a more extensive experimental material was collected to allow also the analysis of the F V and F VI spectra (B- and Be-like), which are less well known than the isoelec-

tronic oxygen spectra O IV and O V. In addition, an unpublished wavelength list kindly made available by Edlén provided highly accurate fluorine wavelengths below 200 Å. Since paper 1.2 (F VII) is intended as the first of several spectroscopic studies of fluorine, it contains a rather detailed description of the experimental material and methods of analysis.

For the comparatively simple Li-like ions semi-empirical methods such as the polarization parametrization [37] and the Ritz formula [38] are of great value in describing and analysing the energy level structure. These methods are rather thoroughly discussed in appendix 2.

In the Li-like term system the energies of all $n\ell^2L$ terms with $\ell > 1$ can be accurately described by the polarization formula with only three parameters: the ionization energy and the dipole and quadrupole polarizabilities of the core. Values of these parameters in O VI and F VII are predicted in [36] and the results of our experimental studies show good agreement between calculated and observed wavelengths for transitions between these terms in both ions. See Table 1 in paper 1.1 and Table 2 in paper 1.2.

A more limited study of the He-like O VII and F VIII spectra reported in papers 1.1 and 1.2 respectively, shows that also in this sequence the polarization formula, with theoretical polarization parameters from Dalgarno [39], accurately describes the hydrogenic part of the term system.

The s and p terms in the Li-sequence on the other hand, are described by two three-parameter Ritz formulae (one for each value of ℓ). In F VII, the predictions in [36] of these terms were also tested experimentally and found to be in good agreement with the observations.

In order to complete the description of the $1s^2n\ell^2L$ term system

in F VII only the ionization potential and the fine-structure splittings remain to be discussed. The ionization limit is accurately determined from the fitting procedures used to obtain the polarization- and Ritz parameters, as discussed in appendix 2. For the fine-structure of the $nd\ ^2D$ terms the experiments show perfect agreement with the hydrogenic values. For terms with $\ell > 2$, where the fine-structure could not be resolved experimentally, calculated splittings should thus be very accurate. Finally, for the 2P splitting Edlén [36] suggested the formulae

$$\Delta(np) = \Delta(2p) (n^*(2p)/n^*(np))^3$$

where the effective quantum numbers n^* are obtained from the Ritz formulae. The experimental results for the 3p to 6p terms verify this scaling.

In summary, we note first of all that the experimental results described above verify the high quality and predictive power of the semi-empirical analysis of the Li-sequence given by Edlén [36]. Secondly, it may be said that the experimental analysis of F VII, among others [40], shows that the results obtained by the beam-foil method are now accurate enough to make a significant contribution in the field of term analysis.

Paper 1.3.

Paper 1.3 reports a study of the transitions $1s2s2p\ ^2\ ^5P - 1s2p\ ^3\ ^5S^o$ in Be-like F VI and $1s2s2p\ ^4P^o - 1s2p\ ^2\ ^4P$ in Li-like F VII.

The resolved $^4P^o - ^4P$ multiplet in the Li isoelectronic sequence has been studied in Li I by Levitt and Feldman [41], in C IV-O VI by Livingston and Berry [42], in F VII (paper 1.3), in Mg X by Träbert et al.[43] and in Al XI by Träbert et al.[44]. The measurement in Li I was performed with the so called Zeeman-quenching atomic-beam method whereas all other studies have utilized the unique property of the beam-foil source to provide a large population

of multiply excited states. The fine- and hyperfine structure in the $1s2p^2\ ^4P$ term in Li I has also been studied by Gaupp et al. [45] using the zero-field quantum-beat technique after beam-foil excitation

The interest in determining accurate experimental wavelengths and fine-structure splittings in this multiplet arises because these systems are important testing grounds for very accurate calculations in three-electron systems. Most of the recent calculations are based on the multiconfiguration Dirac-Fock (MCDF) method [46-48] whereas Chung [49] used a relativistic perturbation technique in order to take better account of correlation effects influencing the transition wavelengths. To obtain accurate fine-structure splittings a detailed calculation of spin-other-orbit and spin-spin interactions must be performed. Hata and Grant [48] also considered radiative (QED) effects, although in F VII they amount to less than 2 cm^{-1} in the fine-structures.

In paper 1.3 a comparison is made between the experimental results in F and the two most recent calculations available at the time [46,47]. This shows that the more accurate calculation of the retardation term in the Breit-operator made by Chen [47] results in a somewhat better agreement concerning fine-structure splittings but an almost equally large (7 Å), but opposite, deviation for the $\lambda_{c.g.}$. After the publication of paper 1.3 two new theoretical studies have appeared [48,49]. While they give approximately the same agreement with experiment as [47] for the fine-structure intervals, they represent a significant improvement in the calculation of $\lambda_{c.g.}$, $+2\text{ Å}$ [48] and -0.17 Å [49] in F VII. The excellent agreement between $\lambda_{c.g.}$ calculated by Chung [49] and experiment prevails throughout the observed part of the sequence showing the importance of accurately including the correlation effects.

The experimental identification of the $^4P^o - ^4P$ multiplet in F VII was complicated by the large number of unknown lines around 820 Å, where the multiplet was predicted to appear. Thus, both $2sn\ell - 2pn\ell$ and second order $2p3\ell - 2p4\ell'$ transitions in F VI are expected in the same wavelength region. The continued analysis of the F VI spectrum has, however, led to the identification of some of these lines. In particular the rather strong line at 820.36 Å has been identified as the $2s3s\ ^1S - 2p3s\ ^1P$ transition. Unfortunately the stronger 817 Å line is still unidentified.

Paper 1.4.

Paper 1.4 reports an extension of the work on the low configurations $3d^3$, $3d^24s$ and $4p$ in Cr IV by Ekberg [50], to include also $3d^24d$ and $5s$.

In this study the light source was a low-voltage sliding-spark discharge, and the spectra were recorded by Ekberg at the 10.7 m normal incidence spectrograph at NBS and at the corresponding 3 m instrument at our institute.

The analysis has primarily resulted in the identification of 59 $3d^24d$ and 15 $3d^25s$ levels based on 391 observed combinations with $3d^24p$ in the 900 - 1800 Å interval. In addition, about 100 new $3d^3 - 3d^24p$ and $3d^24s - 3d^24p$ transitions have been found and the whole term system $3d^3$, $3d^24s, 4p, 4d$ and $5s$ has been recalculated using the combined material from this investigation and the one reported in [50]. Thus, paper 1.4 gives 741 Cr IV lines between 500 and 2500 Å and 154 of the 163 possible energy levels in the above mentioned configurations.

It is interesting to note that also in this case the simple quantum-defect formalism, described in appendix 2, can be used to estimate the ionization limit, although the Cr IV term system is of course much more complicated than the Li-like one discussed

above. The $3d^2(3F)ns$ series is used, but since only two members ($n = 4$ and 5) have been observed the full Ritz treatment can not be used. Instead we proceeded in the following way. The term value was defined as

$$T = E_1 + E(3d^2 3F_{c.g.}) - E(3d^2(3F)ns^2 4F_{c.g.}).$$

The ionization potential is known in the isoelectronic systems Ti II [51] and V III [52], thus the difference in the effective quantum numbers for $4s$ and $5s$ could be determined and extrapolated to Cr IV. Combining this with the formula $T = R \zeta^2 / (n^*)^2$ leads to an equation that can be solved numerically for E_1 .

6. STUDIES OF ATOMIC LIFETIMES.

6.1 ANALYSES NOT REQUIRING EXTENSIVE CASCADE CORRECTIONS.

The division of this section into two parts has no real physical significance and is only done in order to structure this thesis. However, it may also help to emphasize, that despite the extensive discussion of cascades in the literature, there are many situations where this is not a serious problem.

Paper 2.1.1.

Paper 2.1.1. reports lifetime measurements of the levels $2p$ $^2P_{1/2\ 3/2}$ in Li-like Cl XV, $2p^2$ 1D in Be-like Cl XIV and $2p^3$ $^2D_{5/2}$ in B-like Cl XIII. The experiments were done at high energies (40-50 MeV) using the 6.7 MV EN tandem accelerator at the Tandem Accelerator Laboratory in Uppsala.

Analysis of the recorded intensity decays, using curve-fitting techniques, was simple in all but one case. This is because the direct cascades to these low-lying levels, have very short lifetimes and thus affect only the beginning of the observed decay curves. As an example, the analysis of the $2p$ $^2P_{3/2}$ decay resulted in a two-exponential fit giving the primary lifetime 0.71 ± 0.03 ns and a growing-in component of 0.06 ns which then represents some "average" of the many short-lived cascades. The decay of the $2p$ $^2P_{1/2}$ (Cl XV) and $2p^3$ $^2D_{5/2}$ (Cl XIII) required some extra consideration since the transitions at 416 and 419 Å to $2s$ $^2S_{1/2}$ and $2s2p^2$ $^2P_{3/2}$ respectively could not be resolved. However, the lifetimes of these levels are different by a factor of 7 and thus we attributed the short component of a two-exponential fit to the lifetime of the 2D level whereas the long one was taken to represent the 2P level.

A comparison of the observed lifetimes with theoretical predic-

tions showed good agreement in all cases. Our experimental lifetimes were also converted to oscillator strengths, using theoretical branching ratios when necessary. In this respect it is interesting to note that the $2p^2\ ^1D_2$ level has a non-negligible transition probability for the spin-forbidden combinations with $2s2p\ ^3P_{1,2}$ [53]. This was taken into account in the f-value calculation.

Papers 2.1.2 - 2.1.4 all deal with the determination of transition probabilities for so called forbidden transitions, i.e. transitions that are not allowed in the pure LS-coupling scheme. We will show that careful lifetime measurements can provide information on e.g. spin-forbidden combinations (paper 2.1.2 and 2.1.4) and hyperfine induced decay modes (paper 2.1.3). In combination with a theoretical study, experimental information on these transition probabilities provides a powerful aid in probing atomic structure (see appendix 3). Intercombination lines are also of great astrophysical and plasma physical interest as discussed by Gabriel and Jordan [54].

The principle behind the measurements is very simple. In pure LS coupling it may be shown (see appendix 3) that, neglecting a small wavelength factor, all fine-structure levels of a term have the same lifetimes, i.e the same sum of transition probabilities. However, if one of the levels has an additional decay channel, not allowed in LS-coupling, its lifetime will be shorter than for the other fine-structure levels. The "extra" transition probability can then be obtained directly by subtracting the observed decay rates. However, a very high experimental accuracy is needed since the result is obtained as a difference between observed quantities, each with its own uncertainty.

Paper 2.1.2.

In paper 2.1.2 we study the decay of the $2s3p\ ^3P_{0\ 1\ 2}$ fine-structure levels in Be-like N, O and F. In this case the spin-orbit interaction causes mixing of the $3p\ ^1P_1$ and 3P_1 levels which opens additional decay channels for the 3P_1 level, primarily the $2s^2\ ^1S_0$ - $2s3p\ ^3P_1$ intercombination line.

Due to the mixing of the $2s3p$ and $2p3s$ configurations the 3P_1 level can also decay to $2p^2\ ^1S$ and 1D . A theoretical calculation showed, however, that these channels only contribute a few percent to the total intercombination probability. This conclusion was later corroborated in a more detailed theoretical study by Hibbert [55] who, for example, found that the branch $2p^2\ ^1D_2 - 2s3p\ ^3P_1$ amounted to 10% in O V.

The lifetime measurements were performed in the $3s\ ^3S_1 - 3p\ ^3P_J$ decay channels and resulted in a significant difference between the lifetime of 3P_1 and that of $^3P_{0\ 2}$ in all ions, although the effect was rather small in N IV. Assuming that this difference was due entirely to the $2s^2\ ^1S_0 - 2s3p\ ^3P_1$ transition, its probability could be determined as discussed above.

Previous theoretical studies of intercombination lines in the Be I isoelectronic sequence had mainly been concentrated on the intercombination transition from $2p\ ^3P_1$, where the effect is far too small to be measured with this technique, except for very highly ionized atoms [56]. For low-Z ions, only Odabasi [57] had considered the $2s^2\ ^1S_0 - 2s3p\ ^3P_1$ intercombination line but his theoretical values are found to be too high by roughly 50, 60 and 100% in N IV, O V and F VI respectively. In that study only configuration mixing between the odd configurations (9 in number) was considered which might explain part of the disagreement.

The publication of paper 2.1.2 inspired a renewed interest in

this intercombination line. As already mentioned, Hibbert [55] has performed an accurate CI-calculation for N IV - Ne VII obtaining good agreement with our experimental transition probabilities in N IV - F VI. His prediction in Ne VII was subsequently tested experimentally by Hardis et al.[58] who, however, obtained an intercombination probability of $(430 \pm 60) \cdot 10^6 \text{ s}^{-1}$ in disagreement with the calculated value of $275 \cdot 10^6$. The Ne VII - Fe XXIII interval was then studied by Ellis [59] using multiconfiguration wavefunctions and treating relativistic effects as perturbations. His resulting transition probabilities are in good agreement with the experimental value for Ne VII [58], and with the theoretical results of Sampson et al.[60], for ions with $Z > 19$. In the intermediate Z-region, however, the different theoretical studies give drastically different results, as shown in Fig 5, and additional experimental studies are clearly called for to clarify the situation.

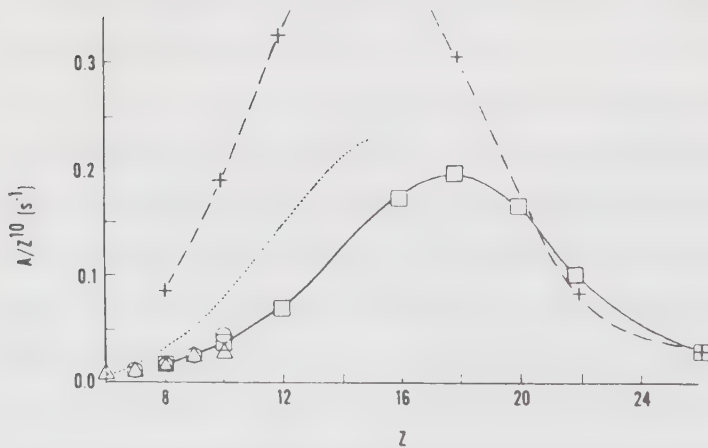


Fig 5. Scaled theoretical transition probability for the intercombination transition $2s^2 \ ^1S_0 - 2s3p \ ^3P_1$. Solid line (squares) Ellis [59], dotted line Odabasi [57] and dashed line (crosses) Sampson [60]. (Figure taken from Ref. 59).

As a continuation of the study of intercombination lines we have also started to investigate the decay-properties of the analogous levels $3s4p\ ^3P$ in Mg-like S V and Cl VI [61].

Papers 2.1.3 and 2.1.4.

Papers 2.1.3 and 2.1.4 report a study similar to the one described above, of the $1s2p\ ^3P_{0\ 1\ 2}$ levels in He-like O VII and F VIII.

For these levels in the He-sequence several decay modes are of importance. In addition to the E1 transition to $2s\ ^3S_1$, allowed for all the fine-structure levels, 3P_2 can also decay to the ground state by a M2 transition. This probability is, however, small in O VII and F VIII [62], but becomes important at higher Z.

Just as in the Be-sequence the 3P_1 level has an additional decay channel through the intercombination line $1s^2\ ^1S_0 - 1s2p\ ^3P_1$. Contrary to the Be case, however, this decay mode is the dominant one already at $Z = 8$.

In O VII the 3P_0 level has only one decay channel, to $2s\ ^3S_1$, since $J = 0$ to $J = 0$ transitions are strictly forbidden. In F VIII on the other hand, where the nucleus has a spin ($I=1/2$) the hyperfine interaction causes a mixing of the $F = 1/2$ states of 1P_1 , 3P_1 and 3P_0 thus opening the $1s^2\ ^1S_0(F=1/2) - 1s2p\ ^3P_0(F=1/2)$ decay mode. The same applies also to the $^3P_2(F=3/2)$ state but since the separation of 3P_1 and 3P_2 is about 7 times that of 3P_0 and 3P_1 [63] this effect is much weaker. Hfs-effects have, however, been observed for 3P_2 in V XXII by Gould et al. [64].

A comparison of our experimental lifetimes in O and F with theoretical results shows good agreement with the relativistic calculations, for example Lin et al.[65], for both 3P_1 and 3P_2 . If the non-relativistic f-values, for example Schiff et al.[66], are combined with the non-relativistic wavelength for the $2s\ ^3S -$

$2p\ ^3P$ multiplet, we obtain 3P_2 lifetimes outside the experimental uncertainties. However, if the experimental transition energy is used instead a reasonable agreement is obtained, thus, corroborating the conclusion of Schiff et al.[66] that the non-relativistic f-values are rather accurate and that relativistic effects mainly influence the energy separation.

For the 3P_0 level in F VIII we obtain a lifetime significantly lower than that predicted by theory. This is due to the hyperfine-induced decay to the ground state. Such effects in the He-sequence have been studied theoretically by Mohr [67] and experimentally in Al XII by Denne et al.[68] and in P XIV by Livingston and Hinterlong [69].

In order to deduce the hyperfine-induced transition probability we subtracted the experimental decay rate from 3P_0 with that from 3P_2 . The latter one was taken to represent the allowed $2s - 2p$ probability after first correcting for the small M2 probability of the $1s^2\ ^1S_0 - 1s2p\ ^3P_2$ channel (less than 1%) and then taking the σ^3 dependence of the lifetimes, discussed in appendix 3, into account. The last correction is around 5%. The value thus obtained $(1.42 \pm 0.2) \cdot 10^7\ s^{-1}$ is in excellent agreement with Mohrs calculated value of $1.3 \cdot 10^7\ s^{-1}$.

Fig 6 shows a comparison between the experimentally determined 3P_0 lifetimes and the theoretical results of Mohr [67]. It is obvious that further experiments are needed in order to test the large effect predicted at higher Z-values

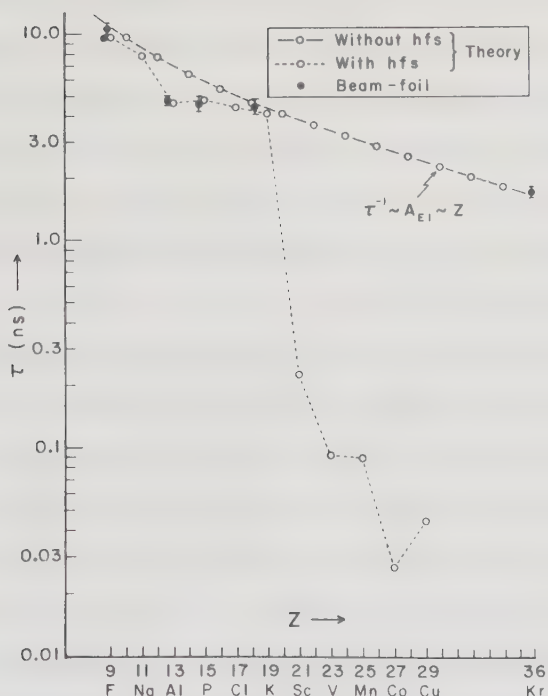


Fig 6. Lifetimes of the $2p\ ^3P_0$ level in He-like ions with and without the hyperfine induced effect. (Figure taken from Ref. 69).

6.2 LIFETIME ANALYSES INVOLVING CASCADE CORRECTIONS THROUGH THE ANDC METHOD

As the previous section illustrated, there are many highly interesting situations where the analysis of the measured decay curves do not present any particular problems and where the lifetimes can be accurately determined from a careful curve-fitting analysis. Common to all these cases was the fact that the (ever present) cascading levels had very different (shorter) lifetimes compared to the levels under investigation.

When this is not so, the sum of exponentials describing the decay will contain terms that are not too different. It should be intuitively clear that a numerical fit may then give a primary lifetime representing some sort of weighted average value and/or unstable solutions where it is difficult to decide on the best number of exponentials to use. This point is easily substantiated by analysing simulated decay curves and several examples will be discussed below.

As an additional example of the problems that may occur, we note that in a complex cascade situation it is possible to encounter cancellation effects. That is, the combination of lifetimes and relative level populations can be such that the exponential term containing the primary lifetime is suppressed. This was pointed out by Curtis [70], who also gives an example of a three-level decay-system (lifetimes: 1,2 and 5 and relative populations: 5,6 and 5) where the primary lifetime component is completely missing. It is of course obvious that this represents an impossible situation for curve-fitting analysis.

Several modifications of the curve-fitting technique have been used in order to reduce the problems of cascades. They include, fits to the numerically differentiated or integrated observed decay curves (to enhance the fast and slow decaying components, respectively) and various types of constrained fits. These methods are thoroughly discussed by Curtis [70] and briefly reviewed in the introduction in paper 2.2.1 where references to some recent work are also given.

A completely different approach is the ANDC technique, suggested by Curtis et al.[71] in 1971. This method utilizes only experimental information and accounts, in principle, exactly for cascades. The general idea is to measure experimentally all (important)

direct cascades to the primary level and then use this additional information to obtain an accurate primary lifetime. The beauty of this scheme is that the direct cascades contain the whole contribution from the rest of the term system. Of course, in a practical application only the most important direct cascades can be measured and included in the analysis, but still, these will contain a large amount of information.

The mathematical formulation of the ANDC technique is discussed in [70,71] and also in papers 2.2.1 and 3.4 and will not be repeated here. Paper 2.2.1 deals with the practical implementation of the ANDC principle, and in the following two papers this method is used to obtain accurate lifetimes in Be-like N IV and O V (paper 2.2.2) and Na-like S VI (paper 2.2.3).

Paper 2.2.1.

Paper 2.2.1 reports a detailed study on how best to implement the ANDC principle in practice, i.e. which mathematical formulation and raw-data treatment to use. The computer program CANDY resulting from these studies is then extensively applied to simulated decay-curves both to test the program and to establish criteria for the validity of an ANDC result in a general case.

The single most important result from these investigations is to show the numerical power and experimental convenience of using a multi-exponential function as a global smoothing before attempting the ANDC analysis. In view of the discussion above of the inadequacy of a multi-exponential fit, this result might at first seem like a paradox. However, in CANDY the multi-exponential functions are only used for smoothing purposes and not to obtain any lifetime parameters, and this functional form is still the most correct representation of a decay curve.

It is interesting to briefly comment the results obtained both

by CANDY and by the multi-exponential fitting program DISCRETE [72] for the simulated decay-data (Table 1 and section 5 in paper 2.2.1), since analyses of synthetic data is the only way to unambiguously test the different methods.

As an example of the strength of the ANDC principle, and also to show that the results of CANDY does not depend on the actual values of the lifetimes in the smoothing functions, the first simulation is the "pathological" cascade scheme discussed above, where the primary decay component is made to disappear. However, CANDY still reveals the correct lifetime since its value manifests itself in the relative admixtures of the direct and indirect cascades.

The last three entries in Table 1 (paper 2.2.1) represent very difficult but realistic situations (Cu-like Mo XIV, K-like Ti IV and Be-like O V respectively) including a large number of cascades, several of which have lifetimes very close to those of the primary levels. The results show that an ANDC analysis performed with CANDY does give primary lifetimes that are correct within the estimated uncertainties. The very sophisticated multi-exponential fitting program DISCRETE on the other hand, is seen to overestimate the lifetimes, although by no more than 10 - 20% typically.

These simulations are then also used to establish criteria by which to judge the quality of an ANDC result. The general problem is, however, so complicated that no exact and simple criterion has been found. Instead, a list containing 7 different test conditions is given in paper 2.2.1, and the analyses of the synthetic data show that together these are very powerful in revealing e.g. missing important cascades, erroneous background subtractions or the use of bad multi-exponential smoothing functions.

The next two papers describe the application of CANDY to real physical situations.

Papers 2.2.2 and 2.2.3.

Paper 2.2.2 reports lifetime measurements of the $2p\ ^1P$ level in Be-like N IV and O V whereas in paper 2.2.3 the lifetimes of several levels in Na-like S VI are given.

Lifetimes and oscillator strengths in the Be I and Na I iso-electronic sequences have been extensively studied both experimentally and theoretically, in particular for the lowest levels. A comparison between the experimental f -values for the resonance lines, obtained by the beam-foil method, and the theoretical calculations show that the former values are systematically somewhat lower in many ions [73,74].

Since many different elaborate theoretical methods have been used, with almost identical results, it has been suggested that the discrepancy with the experimental results is due to an incomplete correction for cascades in the analyses of the beam-foil data.

In the Be-sequence it is possible to be more specific and pinpoint the $2p^2\ ^1S$ level as the probable cause for these difficulties. First of all the lifetimes of the $2p\ ^1P$ and $2p^2\ ^1S$ levels are very similar throughout the sequence, as pointed out by Pegg et al.[75]. Secondly in Be I where $2p^2\ ^1S$ lies above the ionization limit with a high probability of autoionization, an excellent agreement on the $2p\ ^1P$ lifetime is obtained between the beam-foil result of Martinson et al.[76] and the theoretical data e.g. Ref. 77.

The main intension of our studies in these sequences was to test these suggestions and to take up the challenge from the theoreticians and show that accurate experimental lifetimes can indeed be obtained with the ANDC method of analysis.

Analyses of the $2p\ ^1P$ decay curve in O V with the multi-

exponential fitting program DISCRETE [72] resulted in a stable decomposition into five exponentials. Disregarding the uncertain fifth component, all other lifetimes could be directly and rather accurately related to the levels $2p\ ^1P$, $3d\ ^1D$ and $2p^2\ ^1S$ and 1D . Results of similar quality were also obtained in N IV. (See section 3.1 in paper 2.2.2). These analyses are probably among the best curve-fitting results ever reported, and illustrate the high performance of the program DISCRETE.

However, the $2p\ ^1P$ lifetimes were still some 15% longer than those obtained in the subsequent ANDC analyses performed with CANDY. (Thus, in the words of Tom Lehrer from his song New Math: "... the important thing is to understand what you are doing, rather than to get the right answer..." [78]). Since the ANDC results are in excellent agreement with the theoretical lifetimes this verifies the initial suspicion that the discrepancy between theory and experiment was indeed due to problems of cascades in the earlier beam-foil investigations.

In the study of Na-like S VI we measured the lifetimes of all levels in the yrast chain from $3p$ to $8k$. These levels cascade into each other and that repopulation channel is by far the most important one. Thus, by successively applying the ANDC technique we obtained accurate lifetimes for all the yrast levels up to $7i$. In the analyses of the $3p\ ^2P$ levels the cascades from $4s$ and $4d$ were also included, although they had only a marginal effect on the ANDC analyses.

Also in this case we did obtain qualitatively reasonable curve-fitting results, in good agreement with previous experiments for most levels, but 10 - 20% longer than predicted by theory. Cascade corrections with CANDY bring, however, once again theory and experiment into good agreement.

The experimental material and ANDC analyses in S VI discussed here are also used in paper 3.4 to derive accurate relative level populations in S VI. This is discussed in the next section.

In our effort to determine accurate lifetimes in multiply ionized atoms with the ANDC technique we have also studied the $3p^2P$ levels in Na-like Cl VII [79] and an investigation of the $3p^1P$ level in Mg-like S V and Cl VII is under way [80].

To summarize this section we may state that extensive tests, on both synthetic and real data, have shown that the ANDC method, as it is implemented in CANDY, is capable of giving accurate lifetimes also in very difficult situations.

7. STUDIES OF THE BEAM-FOIL EXCITATION MECHANISM.

A study of the beam-foil interaction mechanism is of course of considerable interest from a beam-foil-experimentalists point of view, but also as one way to study the general problem of the penetration of heavy particles through matter. The interaction between the beam ions and the foil manifests itself in many ways. Those of particular relevance in a beam-foil experiment are:

1. Charge state distributions in the emerging beam.
2. Energy loss, energy straggling and angular scattering of beam particles.
3. Polarization of the light emitted by the excited ions.
4. Relative population of excited states within a given charge state.

A comprehensive study of the beam-foil interaction is thus seen to require both optical spectroscopy and other techniques. A brief review of such work prior to 1977 is given by Berry in Ref.13. For a discussion of recent results concerning the first two points, which are intensively studied within the field of collision physics, the reader is referred to e.g. [81]. However, there seems to be no review articles that cover all of the above listed effects.

The papers in this section report experimental results concerning the last two points above. Before discussing them, however, let us briefly comment on the first two, although not as probes of the interaction mechanism (that would be beyond the scope of this thesis) but only as to their influence on a beam-foil experiment.

Charge state distributions have already been discussed in section 3 and references to calculations and tabulations of such distribu-

tions are found in [18]. When a fast ion passes through matter some of its energy is lost due to inelastic interactions with the target atoms. This energy loss must be taken into account to obtain an accurate velocity of the ions after the foil in a lifetime measurement, unless this velocity is measured directly, for example with an electrostatic analyser. The energy loss is usually calculated from the electronic (nuclear) stopping power and the thickness of the foil and depends on foil material, beam element and energy. At MeV energies the nuclear stopping power is small and usually neglected. An extensive tabulation of electronic stopping powers has been done by Northcliffe and Schilling [82], and Garnir-Moujoie et al. [83] have derived simple semi-empirical formulae to calculate both electronic and nuclear stopping powers for a carbon target.

Energy straggling and angular scattering both lead to an energy dispersion in the beam which influences a lifetime measurement since it destroys the exact correspondance between distance from foil and time after excitation. However, Curtis et al.[84] and Kay [85] have shown that this effect is very small. The angular dispersion of the beam results from multiple scattering processes in the foil. Högberg et al.[86] give a simple formula to calculate the angle, $\theta_{1/2}$ (in degrees), corresponding to the half-width of this distribution after a carbon foil:

$$\theta_{1/2} = 12 Z^{3/4} t / E$$

where Z is the atomic number of the projectile element, t the foil thickness in $\mu\text{g}/\text{cm}^2$ and E the energy in keV. This formula was found to fit experimental data for a number of ions at low energies (< 100 keV) and thin foils [86]. A general theoretical study of the angular distributions is given by Sigmund and Winterbon [87] and their results seem to be in excellent agreement with experiment [88].

The effect of the angular scattering, at MeV energies, is mainly to limit the resolving power obtainable in a spectroscopic study, since the induced line-of-sight velocities lead to a Doppler broadening of the spectral lines. As a numerical example (of relevance at our normal energies) we can consider a 5 MeV F-beam and a $10 \text{ } \mu\text{g}/\text{cm}^2$ carbon foil. Using either the formula above [86] (outside its tested energy range) or the results of [87] we obtain $\theta_{1/2} = 0.12^\circ$ and a Doppler effect $\Delta\lambda/\lambda$ of $1.03 \cdot 10^{-4}$ which corresponds e.g. to a spectral line width (FWHM) of $0.2 \text{ } \text{\AA}$ at $2000 \text{ } \text{\AA}$.

Paper 3.1.

We now turn to the papers of this section. Paper 3.1 reports the observation of a high degree of circular polarization of the light emitted by 4 MeV oxygen ions after excitation in tilted foils.

In the normal beam-foil geometry, i.e. with the foil perpendicular to the beam, the cylindrical symmetry of the excitation prohibits the formation of oriented atomic states and hence the observation of circular polarized light [13,89]. In the experiment we studied the spectral lines 3433 and 3737 $\text{\AA}$. The former is a blend of 6fgh - 7ghi transitions in O VI and the latter is the $2p3p \text{ } ^4\text{D}$ - $2p3d \text{ } ^4\text{F}$ line in O IV. The degree of circular polarization was studied as a function of foil tilt-angle between 0° and 50°

In both cases the amount of circular polarization was found to increase linearly with tilt-angle, reaching 30% for the 3433 $\text{\AA}$ line and about half as much for the 3737 line at an angle of 50° . The results for the 3433 $\text{\AA}$ line in O VI were later corroborated in an extended study [90] using a more sophisticated experimental technique [91,92]. These results show that the exit surface of the foil plays an important role in the beam-foil excitation mechanism.

The influence of the exit surface on the anisotropy of excited He atoms has been investigated in detail by Winter [93], and a more

systematic use of the state of polarization of the emitted light as a probe of the beam-foil excitation mechanism has been reported e.g. by Schectman et al.[94].

The next three papers (3.2 - 3.4) all report studies of the relative population of excited states after the beam-foil interaction. Such information is, in principle, easily extracted from an observed spectrum. The intensity, I , of a line is given by $I = E(\lambda) A N$, where $E(\lambda)$ is the (relative) efficiency of the detection system at wavelength λ , A the transition probability of the observed decay branch and N the population of the upper level. Thus, if E and A are known for two transitions the relative level population of the corresponding upper levels can be directly established.

The relative efficiency of the monochromator and detector as a function of wavelength is usually determined by observing light emitted by some standard source, e.g. a calibrated deuterium or tungsten lamp. For short wavelengths ($\lambda < 1200 \text{ \AA}$) the only available standard source is synchrotron radiation. A more detailed discussion of different calibration procedures are given by Andersen et al. [95].

As mentioned earlier, there exists no comprehensive theory to predict the distribution of the population on the excited states in the beam after the foil passage. Generally speaking the beam-foil interaction can be divided into two parts, one taking place inside the foil and the other at (or close to) the exit surface. The distance between two carbon atoms in the foil is approximately $2 \text{ \AA}$ and thus, an excited state with a mean radius larger than $1 \text{ \AA}$ can not survive over a longer distance inside the foil. Instead such states are believed to be predominantly populated by electron pickup processes as the ion leaves the foil [96].

Inside the foil, collisions between the beam and the target atoms lead to a complicated balance between ionization, excitation and recombination processes. Ions ending up in geometrically small excited states can then emerge from the foil. A subsequent capture of an electron leads to the formation of a doubly excited state, which can be observed spectroscopically if it is stable against auto-ionization. If not, the very rapid autoionization process ($\sim 10^{-15}$ s) leads to a rearrangement of the electrons into an excited state of the next ionization stage. This superficial description may serve to indicate the complexity of the beam-foil excitation process. It also suggests that one should see differences in the behaviour of the populations of high- and low-lying excited states as the experimental conditions are varied.

Paper 3.2.

In paper 3.2 we report a study of relative level populations in the Li-like ions C IV - F VII. The investigation is concentrated on levels with high n and ℓ quantum numbers, i.e. orbitals with such large radii that they must be populated at, or close to, the exit surface of the foil. Previous experimental studies had shown that for neutral and weakly ionized atoms the relative populations within a given Rydberg series decrease as $(n^*)^{-p}$, where p usually is close to 3 [97].

In C IV and N V we observed the same decreasing behaviour of the population in levels with fixed value of ℓ as a function of n . However, for both O VI and F VII the population curves exhibit broad maxima at n -values around 7 and 8 in O VI and F VII respectively. Also the data for N V can be said to indicate a maximum, in this case around $n = 6$. It should be noted, however, that for the hydrogenic levels with $n > 7$ only $\Delta n > 1$ transitions could be observed.

Thus, the populations shown in Fig 1 (paper 3.2) for these n -values do not include the terms with maximum ℓ . Since the populations are usually found to increase with ℓ (for fixed n), this means that for a consistent comparison of populations for different n -values these curves should be viewed as less steeply descending.

These results are explained as a near-resonant charge transfer from the valence band of the carbon foil. The binding energy of an electron in the valence band of carbon is approximately 10 eV which coincides with the binding energy of the $n = 6, 7$ and 8 levels in N V, O VI and F VII respectively. The levels studied in [97] all had much lower binding energies and there is no conflict with the results reported here. These results support the assumption that the population of the high n, ℓ states is caused by electron capture.

The fact that at least two different excitation mechanisms are active was clearly established in an investigation of relative populations in Na-like Ar VIII by Bashkin et al.[98]. That study included also low-lying levels, exclusively decaying in the XUV region, and showed a large preferential population of the $3p$ and $3d$ levels in addition to population maxima for the high-lying levels at around $n = 11$, corresponding to a binding energy of around 10 eV. These results were interpreted as showing the presence of two different excitation mechanisms [98]. The high-lying levels are populated by the electron-pickup process discussed above, whereas the $3p$ and $3d$ levels are populated mainly through inner-shell, molecular-orbital (MO), electron-promotion processes [99].

Paper 3.3.

Paper 3.3 is a continuation and extension of the Ar VIII work to chlorine. According to the formula $I = E A N$ above, accurate transition probabilities are needed to determine relative level populations.

For the Cl spectra considered here, Cl V - Cl VIII, extensive sets of such data are only available for Na-like Cl VII, from the numerical Coulomb approximation calculations of Lindgård and Nielsen [100]. Thus, for the other ions only the changes in the relative populations as functions of beam energy could be determined.

An extensive discussion of the results is given in paper 3.3 and only two points will be considered here.

The Cl VII results show a qualitative agreement with the findings [98] in the isoelectronic system Ar VIII, mentioned above. We will come back to a discussion of the populations in Na-like ions in connection with paper 3.4.

In view of the discussion of the excitation mechanism presented so far in this section, it is interesting to comment on the excitation functions obtained in paper 3.3 for the 3p3d levels in Cl VI. For each of the levels in the 3p3d configuration the population, as a function of incident beam energy, shows a double-maximum structure (Fig 3 in paper 3.3). This is interpreted as evidence for the presence of several simultaneous excitation mechanisms. At low energy, the probability of a MO process is relatively small and thus, in this energy domain the population comes mainly from capture of two electrons from the foil. However, at increasing projectile energy there will be an increasing probability for some of the projectiles, initially stripped to $7+$, to have experienced a violent collision inside the foil resulting in a 3p or 3d MO electron-transfer process, so that the projectile occurs as an excited Cl^{6+} ion when it enters the foil exit region. Then, capture of an additional electron may give rise to the observed doubly excited 3p3d levels in Cl^{5+} . This combination of processes could account for the second maximum observed at higher energies.

Paper 3.4.

Paper 3.4 shows, for the first time, that the ANDC technique can be used to determine relative level populations from the analyses of cascade correlated decay curves.

The method is thoroughly discussed in paper 3.4 and only a brief summary of its advantages will be given here. The initial intensity can be more accurately determined from an analysis of a decay curve than from a spectrum. This is particularly important for transitions from short-lived levels, where the fact that the observed intensity corresponds to an average over a section of the beam, leads to a systematic underestimation of the true intensity at time zero. See Table 2 in paper 3.4 and also [101,102].

The ANDC analysis provides a measure of the relative detection efficiency at the wavelengths of the primary and cascade transitions. However, to determine relative level populations this efficiency need not be evaluated explicitly (see formula 2 in paper 3.4). This is an important simplification which also increases the accuracy of the method compared to analyses using a recorded spectra. For example, decay curves measured with different normalizations could be used without any corrections, and the dependance of the detection efficiency on the state of polarization of the light does not have to be considered.

This technique is then applied to a study of level populations in Na-like S VI. The experimental material for this analysis was the same as that used in paper 2.2.3., and the quality of the ANDC results concerning the lifetimes has already been discussed. The resulting relative level populations are given in Table 3 and Fig 3 in paper 3.4.

In addition to S VI, population data for Na-like ions are available for Cl VII (paper 3.3), Ar VIII [98] and Ti XII [103]. A comparison shows that the preferential population of the 3p and 3d

levels is clearly established in all these ions. However, for the higher-lying levels interesting differences are observed. In both Cl VII and Ar VIII a pronounced population maximum is observed for levels with a binding energy of around 10 eV. In S VI on the other hand, the populations are almost constant although the binding energy of the 6h, 7i and 8k levels is 13.6, 10.0 and 7.7 eV respectively. The same result is also seen in Ti XII [103] where the populations of the 4f, 5g and 6h levels are found to be almost constant, although the binding energies are much larger in this case.

Paper 3.5.

Paper 3.5 reports an initial series of experiments using a molecular CN^+ beam from the Lund Pelletron tandem accelerator. The negatively charged CN ions were produced in a direct extraction duoplasmatron ion source and a sufficient number survived the charge-exchange collisions in the stripper gas as molecules to produce a beam of 100 nA of energy-analysed CN^+ molecules. The current was found to be very sensitive to the pressure of the stripper gas but rather insensitive to the terminal voltage.

When this CN^+ beam passes through the carbon foil most of the molecules break up, and the light emitted downstream of the foil emanates from transitions in various C and N ions, as shown in the spectrum presented in Fig 1 in paper 3.5. No conclusive evidence was found for the presence of molecular lines.

Two sets of experiments were performed using the CN^+ ions. In the first one, the intensity of several C- and N-ion lines were studied as a function of foil thickness. It was found that the intensity of transitions between the $n = 2$ levels was independent of foil thickness in the interval $1.5 - 100 \mu\text{g}/\text{cm}^2$. For transitions where the upper level had a larger principal quantum number ($n = 3$ or 4) the intensity decreased with increasing foil thickness.

With this experimental setup we could alter the internuclear separation of the dissociated C and N ions after the foil by increasing the foil thickness. The separation is caused mainly by the angular scattering in the foil (discussed previously in this section).

To further investigate the effects of the separation between the ions, the next series of experiments included the possibility of inserting a second foil some 5 cm upstream. The intensities of the 2s - 2p and 4f - 5g transitions in C IV and N V were studied with and without this prefoil. All foils had a fixed thickness of $5\mu\text{g}/\text{cm}^{-2}$ in this case. The intensities of the 2s - 2p transitions were found to be insensitive to the number of foils used, whereas the 4f - 5g intensities clearly decreased when the prefoil was inserted. This behaviour is consistent with the results of the first set of measurements.

The qualitative interpretation of these results is that the geometrically larger $n = 3$ or 4 states are much more sensitive to the proximity of the other ion from the molecule than the smaller $n = 2$ states. Further experiments are under way to study the molecular effects in greater detail.

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APPENDIX 1. PARTIAL CONTENTS OF THE SOFTWARE LIBRARY

1. DATA-COLLECTION AND EXPERIMENT CONTROL.^a

- SOX Initiates a beam-foil measurement. Drives motors to start positions and prepares a random-access file for data storage.
- EOX Main experimental control program. Drives motors, reads scalers, presents and saves incoming data and allows instantaneous changes of the parameters used to control the experiment. Contains on-line versions of some of the programs described below for data reduction.
- CHARGE Calculates charge state distributions according to [18], and accelerator parameters to optimize a desired charge state
- REFOCUS Calculates the cam angle in the Minuteman monochromator needed for the wavelength independent refocusing technique described in section 4.

2. DATA ANALYSES.

2.1 SPECTRAL DATA.

- SPECAN Plots spectrum on a graphics terminal and on paper. Performs a fit of a single Gaussian line profile.
- PEAK^b Completely automatic peak-finding routine. Primarily for reasonably well resolved lines.

- SPECG As SPECAN but fits a sum of up to 10 Gaussian line profiles.
- CARATE^C Powerful interactive program for the decomposition of blended lines. Since it became available in 1982 it has replaced SPECG for most applications.
- LSQR Least squares fitting program. Used to obtain an absolute wavelength scale by fitting a polynomial (degree < 10) to known reference lines. Produces wavelength lists in combination with PEAK and CARATE/SPECG.
- RITZ Fits a Ritz formula to experimental energy levels for the determination of the ionization limit and interpolation/extrapolation of missing levels in a term series.
- POLFORM Fits a polarization formula to experimental energy levels for the determination of the ionization limit and interpolation/extrapolation of missing levels in a hydrogenic term system.
- HTRANS Calculates wavelengths and transition probabilities for hydrogenic transitions including the effect of the dipole- and quadrupole polarizabilities of the core.

2.2. LIFETIME ANALYSES.^d

- LIFE Plots decay curves on a graphics terminal and on paper. Performs a fit of one and two exponential decays.

DECFIT^b Highly interactive program to fit up to 4 exponentials plus different backgrounds.

DISCRETE^e Powerful automatic program to fit up to 9 exponentials.

DISCPREP Prepares input data for DISCRETE

CANDY Performs an ANDC analysis of cascade correlated decay curves (paper 2.2.1).

a. For a thorough documentation see Engström and Huldt [28].

b. Written by S.Huldt.

c. L.Carlén [104].

d. For a thorough documentation see Engström [29].

e. Written by S.W.Provencher [72]

APPENDIX 2. DISCUSSION OF THE POLARIZATION AND RITZ FORMULAE.

For non-penetrating orbitals, i.e. orbitals where the ℓ -quantum number of the outer electron is (much) larger than that of the core electrons, the energy of a level $E(n, \ell)$ is given by

$$E(n, \ell) = E_1 - T_H(n, \ell) - \Delta_p(n, \ell)$$

where E_1 is the ionization energy, $T_H(n, \ell)$ the hydrogenic term-value and Δ_p the polarization energy. The last quantity can be written as

$$\Delta_p = \alpha_d R \langle r^{-4} \rangle + \alpha_q R \langle r^{-6} \rangle$$

where α_d and α_q are the dipole- and quadrupole polarizabilities of the core respectively and the expectation values are given as closed expressions in n and ℓ in [37].

According to this model it is thus possible to accurately describe the infinite number of non-penetrating terms with a formula containing only three parameters (E_1 , α_d and α_q). A remarkable simplification! Conversely, given a number of experimentally determined level values, the above formulae can be used to obtain the ionization limit and the polarization parameters through a multiple linear regression analysis. (Computer code POLFORM in appendix 1).

It should be stressed, however, that this model is based on several approximations. Even for "non-penetrating" levels the penetration energy is of course not exactly zero but only much smaller than for penetrating ones. The model also neglects exchange interactions between the core and valence electrons. Both these effects depend on the overlap between the core and valence electron wavefunctions and decrease as n and ℓ of the outer electron increases. In addition, higher multipole terms are neglected and it is assumed that the core distortion follows adiabatically the motion of the valence

electron. Non-adiabatic corrections are discussed by Eissa and Öpik [105] whereas the other approximations are analysed in a number of investigations. A small selection is given in Refs.106-110

These studies show that the value of α_q is much more sensitive to the approximations than that of α_d . Thus, while both parameters are needed to accurately describe the observed hydrogenic part of a term system, a comparison with theoretical values of the dipole and quadrupole polarizabilities usually shows better agreement for α_d than for α_q .

The experimental results obtained in F VII illustrate this discussion. A comparison between the experimental values of $\alpha_d = (1.63 \pm 0.3) \cdot 10^{-3}$ and $\alpha_q = (1.47 \pm 0.2) \cdot 10^{-4}$ and the corresponding theoretical polarizabilities $\alpha_d = 1.61 \cdot 10^{-3}$ and $\alpha_q = 7.68 \cdot 10^{-5}$ [111, 112] shows good agreement for α_d and the expected discrepancy in the values of α_q . An attempt to include penetration effects before fitting the parameters did indeed reduce the value of α_q but only to $1.30 \cdot 10^{-4}$ showing that effects other than penetration also play an important role.

For penetrating orbitals, deviations from the purely hydrogenic values are caused by both penetration and polarization. In this case, a semi-empirical treatment requires a separate study of each spectral series, i.e. for each value of ℓ . The simple formula

$$T = R \zeta^2 / n^2$$

valid for a hydrogen-like ion of net charge ζ is modified by writing

$$T = R \zeta^2 / (n^*)^2 = R \zeta^2 / (n - \delta)^2$$

since penetration and polarization effects increase the term value of a level. In this formula n^* is called the effective quantum number and δ the quantum defect. δ is a function of ℓ but approximately independent of n . According to the Ritz formula [38] δ may be

written as a polynomial

$$\delta = a + bt + ct^2 + \dots$$

where $t = T/R \zeta^2 = (n^*)^{-2}$, thus taking the small n -dependence into account. With $T = E_1 - E$ where E is the experimental energy we note that the above formulae define an implicit relation between E , E_1 and the Ritz parameters $a, b, c, \dots$. For a fixed number of such parameters, these and the ionization limit E_1 can be determined through a non-linear least squares fit to the observed energies. (Computer code RITZ in appendix 1).

This procedure is applied to the s to g series in F VII. The resulting fits are shown in Fig 3 in paper 1.2 and used to obtain an accurate value of the ionization potential (the error estimate is 3 parts in 10^6). Moreover, having established the necessary parameters the formulae can of course be used to obtain new level values in the series

As a final point, it is interesting to ask how such a simple formula ($T = R \zeta^2 / (n - \delta)^2$) can take into account such diverse effects as penetration, relativity and polarization. Expanding the formula for $n \gg \delta$ we obtain

$$T = R \zeta^2 / (n - \delta)^2 = R \zeta^2 (n^{-2} + \delta n^{-3} (2 + O(\delta/n)))$$

This shows that the correction to the hydrogenic value has a n^{-3} dependance which is just the (approximate) n -dependence obtained for all three effects above! See [108] and formulae for Δ_p and relativistic corrections to T_H given in [37].

APPENDIX 3. THEORETICAL DESCRIPTION OF E1-TRANSITION PROBABILITIES.

This appendix summarizes the definitions of transition probability, line strength and oscillator strength. In addition, it is shown that in pure LS-coupling the lifetime of the individual fine-structure levels in a LS term are identical, except for a small wavelength factor.

The treatment follows closely that of Cowan (Ref.1, chapter 14),

The spontaneous emission transition probability per unit time from an excited state $\gamma'J'M'$ to a state γJM is (in the non-relativistic dipole-length formalism) given by

$$a = \frac{64\pi^4 e^2 a_0^2}{3h} \sigma^3 \sum_q |\langle \gamma JM | P_q^{(1)} | \gamma' J' M' \rangle|^2$$

where the tensor operator P is the dipole moment of the atom in units of $-ea_0$.

Applying the Wigner-Eckart theorem to this matrix element we obtain

$$\langle \gamma JM | P_q^{(1)} | \gamma' J' M' \rangle = (-1)^{J-M} \begin{pmatrix} J & 1 & J' \\ -M & q & M' \end{pmatrix} \langle \gamma J || P^{(1)} || \gamma' J' \rangle$$

where the reduced matrix element is independent of M .

The line strength S is now defined as

$$S = |\langle \gamma J || P^{(1)} || \gamma' J' \rangle|^2$$

The Einstein spontaneous emission transition probability of an atom in a specific state $\gamma'J'M'$ is defined as the total probability per unit time of a transition to any one of the states M in the lower level γJ . Thus,

$$\begin{aligned}
 A(\gamma' J' M' \rightarrow \gamma J) &= \sum_M a(\gamma' J' M' \rightarrow \gamma J M) = \\
 &= \frac{64\pi^4 e^2 a_0^2 \sigma^3}{3h} S \sum_{Mq} \left(\begin{matrix} J & 1 & J' \\ -M & q & M' \end{matrix} \right)^2 \\
 &= \frac{64\pi^4 e^2 a_0^3 \sigma^3}{3h} \frac{S}{2J' + 1}
 \end{aligned}$$

where step 2 is just a collection of the above formulae and the third step follows from the summation rules for 3-j symbols (Cowan, eq.5.15). This result proves that A is independent of M' . The weighted transition probability gA becomes

$$gA = (2J' + 1)A = 2.0261 \cdot 10^{-6} \sigma^3 S \quad [s^{-1}]$$

where the wavenumber σ should be in cm^{-1} and S in atomic units. In passing, we may note that the intensity of a spectral line ($J' \rightarrow J$) is proportional to σgA and thus to $\sigma^4 S$ which explains the name line strength for the quantity S .

Another important quantity is the absorption oscillator strength, f , which gives the total probability of absorption from a specific state γJM of the lower level to all states M' of the upper level $\gamma' J'$.

$$f(\gamma JM \rightarrow \gamma' J') = \frac{8\pi^2 m c a_0^2}{3h} \sigma \frac{S}{2J + 1}$$

To prove that the lifetimes of all fine-structure levels in a LS term are identical we show that

$$\tau_{J'}^{-1} = \sum_J A(\gamma' J' \rightarrow \gamma J)$$

is independent of J' . In an LS-coupled basis the matrix element

$D = S^{1/2}$ can be written generally as

$$D = \langle \dots L S J \parallel P^{(1)} \parallel \dots L' S' J' \rangle$$

However, the electric dipole operator $P^{(1)}$ does not involve spin coordinates which means that uncoupling formulae (Cowan, eq. 11.38) can be used to simplify the reduced matrix element. Thus, D can be written

$$D = \delta_{SS'} (-1)^{L+S+J'+1} \sqrt{(2J+1)(2J'+1)} \begin{Bmatrix} L & S & J \\ J' & 1 & L' \end{Bmatrix} \langle \dots L \parallel P^{(1)} \parallel \dots L' \rangle$$

where the last reduced matrix element is independent of J and J' .

Combining the above formulae we obtain

$$\begin{aligned} \tau_{J'}^{-1} &= \sum_J A = \text{const.} \cdot \sum_J \sigma_{J',J}^3 \frac{D^2}{2J'+1} = \\ &= \text{const.} \cdot \sum_J \sigma_{J',J}^3 (2J+1) \begin{Bmatrix} L & S & J \\ J' & 1 & L' \end{Bmatrix}^2 \\ &= \text{const.} \cdot \frac{\sigma_{av}^3}{2L'+1} \end{aligned}$$

In step 3 we have used the fact that the reduced matrix element, in the expression for D , is independent of J . In the next step the sum rules for 6-j symbols (Cowan, eq. 5.31) are used together with the approximation that all lines in the multiplet have the same wavenumber, σ_{av} . Thus, we find that the lifetime is independent of J' . A similar result can also be derived, in the same way, in all other pure coupling schemes.

As an example of how to perform the calculation of S in a general case, i.e. in intermediate coupling, we may consider how the inter-combination line $2s^2 \ ^1S_0 - 2s3p \ ^3P_1$ (in a Be-like ion) arises.

The level that we label $\ ^3P_1$ is not a pure basis state and should more correctly be written

$$\ ^3P_1 = a_1 \ ^3P_1 + a_2 \ ^1P_1 + a_3 \ ^3P'_1 + a_4 \ ^1P'_1 + \dots$$

where the primed basis functions belong to the $2p3s$ configuration.

Likewise, the level labeled $2s^2 \ ^1S_0$ should be written

$$\ ^1S_0 = b_1 \ ^1S_0 + b_2 \ ^1S'_0 + \dots$$

where S' is the $2p^2 \ ^1S_0$ basis function. In both cases the coefficients a_i and b_i are obtained from the diagonalization of the energy matrices, and a_2 and a_4 are non-zero because of the spin-orbit interaction.

$S^{1/2}$ is then given by the matrix product

$$\begin{pmatrix} b_1 & b_2 \end{pmatrix} D \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix}$$

where the dipole matrix D can be written

$$\begin{matrix} & \begin{matrix} \ ^3P_1 & \ ^1P_1 & \ ^3P'_1 & \ ^1P'_1 \end{matrix} \\ \begin{matrix} \ ^1S_0 \\ \ ^1S'_0 \end{matrix} & \begin{pmatrix} 0 & D_1 & 0 & 0 \\ 0 & 0 & 0 & D_2 \end{pmatrix} \end{matrix}$$

with only two non-zero matrix elements. The total matrix product then becomes

$$S^{1/2} = b_1 a_2 D_1 + b_2 a_4 D_2$$

showing the non-zero transition probability for the so called forbidden transition $2s^2 \ ^1S_0 - 2s3p \ ^3P_1$.

This discussion shows also that a comparison with an experimental value for such a transition probability is a very sensitive test on the detailed composition of an atomic state.

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